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Pharos — A Chopper Spectrometer for Inelastic-Neutron-Scattering Studies of Excitations in Materials

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The Pharos spectrometer has been a part of the LANSCE user program for nearly a decade. Originally, the instrument possessed only a low-scattering-angle flight path used primarily for neutron Brillouin scattering studies.[1] However, in the last year, a major upgrade of Pharos was completed, adding 317 new position-sensitive detectors at large scattering angles (for a total of 392), new data-acquisition system hardware and software, a more reliable Fermi chopper system, a new T_0 chopper control system, and a new vacuum system. These upgrades have expanded the scope of scientific problems that Pharos can address and have improved the reliability and efficiency of the instrument. In addition, the suite of Pharos sample environments are presently being expanded to include high temperatures (up to 1500°C), large magnetic fields (up to 11 T), and high pressures (up to 10 kbar). Pharos can now perform a full range of inelastic-neutron-scattering measurements, including phonon and spin-wave dispersions in single crystals; phonon and spin-wave densities of states; crystal-field, spin-orbit, and other electronic excitations; and neutron Brillouin scattering.

Pharos Specifications

Moderator and beam transport. Pharos resides on a 12- x 12-cm^2 ambient high-resolution water moderator gadolinium poisoned at a depth of 1.5 mm. The instrument views the moderator at a 15° angle. The beam is reduced down to a 5- x 7.5-cm^2 beam at the sample position (20 m from the moderator) by continuous beam scraping.

Fermi choppers. Pharos is a direct-geometry Fermi chopper spectrometer. The Fermi chopper monochromates the incident beam by opening a short time window (~ 10 µs) at a certain time delay from the creation of the neutron pulse. Because the neutron velocity and arrival time at the chopper are related simply by the distance from the moderator, the Fermi-chopper time window passes a very narrow energy distribution. The efficiency of transmitting neutrons through the Fermi chopper depends on the chopper rotational frequency, body radius, slit radius, and neutron incident energy. Pharos has three different Fermi choppers that service different incident energies (from ~ 10 to 1,000 meV). These choppers reside at a distance of 18 m from the moderator. Details and performance characteristics of the Pharos Fermi choppers are shown in Table I.

Detector and data-acquisition systems. Pharos is outfitted with 392 1-in.-diam ^{3}He-filled position-sensitive detectors (PSDs). The detector bank

Table 1. Pharos specifications

Energy transfer resolution	ΔE/Ei = 1% to 4%
Moderator	Chilled water at 283 K (1.5-cm Gd-poisoning depth)
Beam size	5 cm x 7.5 cm
Sample distance from moderator	20 m
Fermi chopper	
- Distance from moderator	18 m
- Frequency	60 Hz to 600 Hz
- Diameter	10 cm
- Optimal energy at 600 Hz	100, 300, and 1,000 meV
- Slit spacing	3.6 mm, 2.1 mm, and 1.0 mm
- Slit curvature	0.58 m, 1.0 m, and 1.83 m
- Phasing error to alternating-current line (FWHM)	~ 1.5 µs
- Manufacturer	Revolve Magnetic Bearings Inc.
T_0 chopper	
- Distance from moderator	14 m
- Material	Inconel (341 kg)
- Frequency	10 to 60 Hz
Detectors	
- Distance from moderator	24 m (low angle movable to 30 m)
- 376 position sensitive detectors	1 in. diam., 1 m long
- 16 position sensitive detectors	1 in. diam., 16 in. long, above and below direct beam
- Scattering-angle coverage	1.5° to 145°
- Solid-angle coverage	0.7 sr
- Positional resolution (FWHM)	< 1 cm
- Manufacturer	Reuter-Stokes Inc.

contains 376 1-m-long tubes at a distance of 4 m from the sample. The tubes continuously cover horizontal scattering angles from -10° to -1.5° and 1.5° to 145° and vertical angles from -7° to +7°. The detector bank also contains sixteen 16-in.-long PSDs (above and below the direct beam). The low-angle bank can be moved to 10 m from the sample when smaller scattering angles or higher resolution are required, for example, for neutron Brillouin scattering studies. The entire detector bank covers a solid angle of nearly 0.7 sr. The detectors have position sensitivity better than 1 cm, but they are typically divided into 1-in. elements for data collection. (There are ~ 15,000 detector pixel elements of 1 x 1 in.). The data-acquisition system has a timing resolution of 100 ns, which is too fine for inelastic-neutron-scattering research. We typically work with several thousand time channels of 1 to 3 μs in size over a range of several milliseconds. Thus, the instrument contains an astounding 50 million individual elements to create the histogram. In principle, we can create histograms over the full pulse frame (50 ms), which contains several Fermi chopper transmissions (since the chopper rotates several times faster than the source frequency), each of which has a different incident energy and resolution. This repetition-rate-multiplication technique can be used to increase data rates by 4 or 5 times.[2]

Background suppression. Due to the monochromated beam and the intrinsic weakness of an inelastic-neutron-scattering signal (which is several orders of magnitude less than elastic Bragg scattering), the suppression of extrinsic sources of neutron and gamma background is extremely important. The T_0 chopper attenuates fast neutrons and gamma rays from the prompt pulse by rotating a large Inconel chopper into the beam precisely at T_0. Without the T_0 chopper, fast neutrons would thermalize in the shielding and leak out over time, contributing to a significant background signal, especially at times below 5 ms. The Pharos T_0 chopper can rotate up to 60 Hz in multiples of 10 Hz. To minimize air scattering in the secondary flight path, the entire secondary spectrometer vessel (1,100 m^3) is evacuated down to 10^{-3} Torr. A separate smaller sample chamber can reach cryogenic vacuum levels (10^{-8} Torr). In the near future, we plan to install a radial collimation system in the secondary flight path to reduce the scattering from bulky sample environments (such as the 11-T cryomagnet and gas pressure cells). We find little impact from noise originating in the detector electronics.

Sample environments. Pharos presently supports sample environment equipment for low temperatures down to 2 K (i.e., two displexes and a 70-mm orange cryostat) and high temperatures up to 1,800 K (i.e., a cryofurnace and high-temperature furnace). We are also developing and testing a high-pressure gas cell (to 10 kbars) for the orange cryostat and plan to have an 11-T split-pair superconducting magnet available for users in 2003. Finally, we are collaborating with the Spallation Neutron Source (SNS) and the California Institute of Technology for a prototype two-axis, single-crystal rotation stage that operates at temperatures down to 10 K.

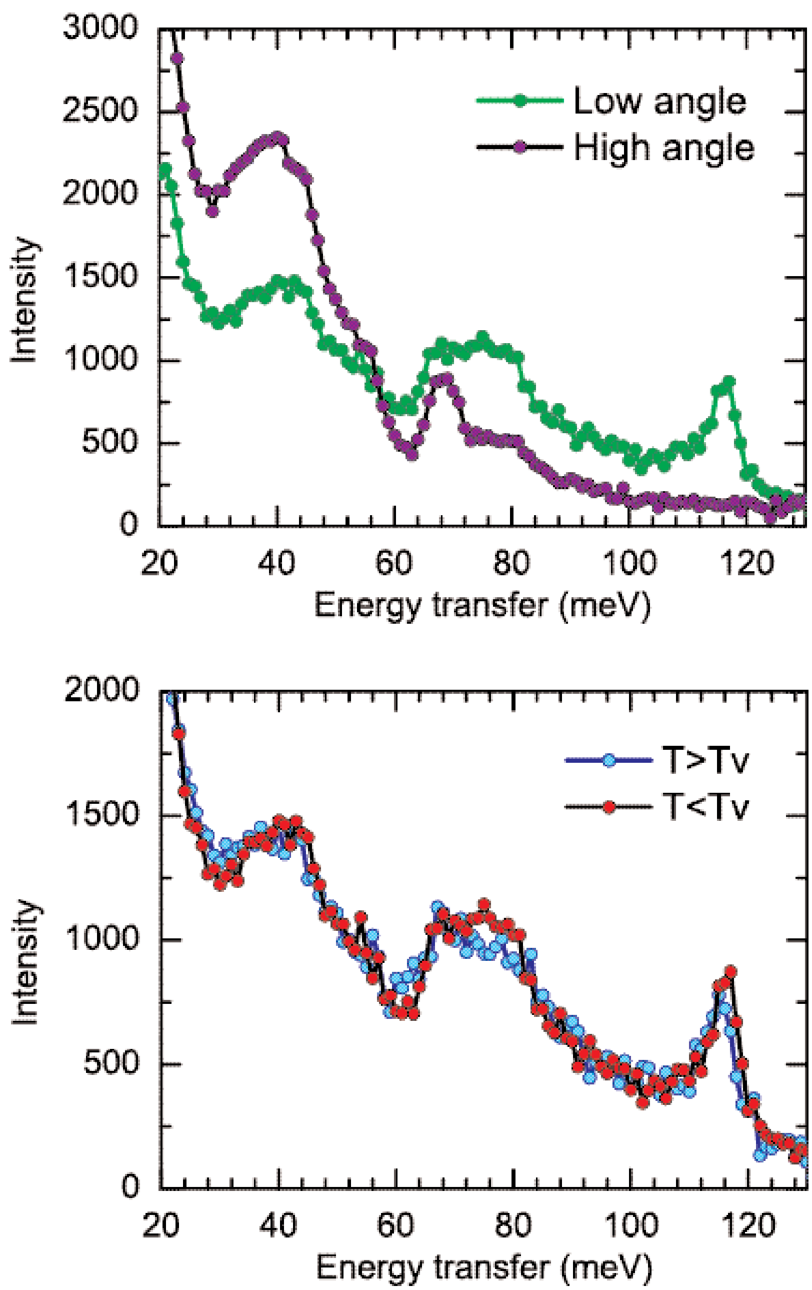
Recent Scientific Results on Pharos

The initial measurements performed on Pharos were scientific-commissioning experiments designed to see how the instrument performs with different samples and for a variety of physics applications.

Lead single-crystal. Phonon measurements were made on a single crystal of lead to determine the viability of performing measurements of phonon-dispersion curves. Although the lead crystal was large (~ 20 cm^3), the phonon energies were low (< 10 meV), showing that Pharos performs quite well in the low-energy region of the spectrum. The experiment was very successful. Data were collected for 9 hours, although dispersion curves were seen after only 1 hour.

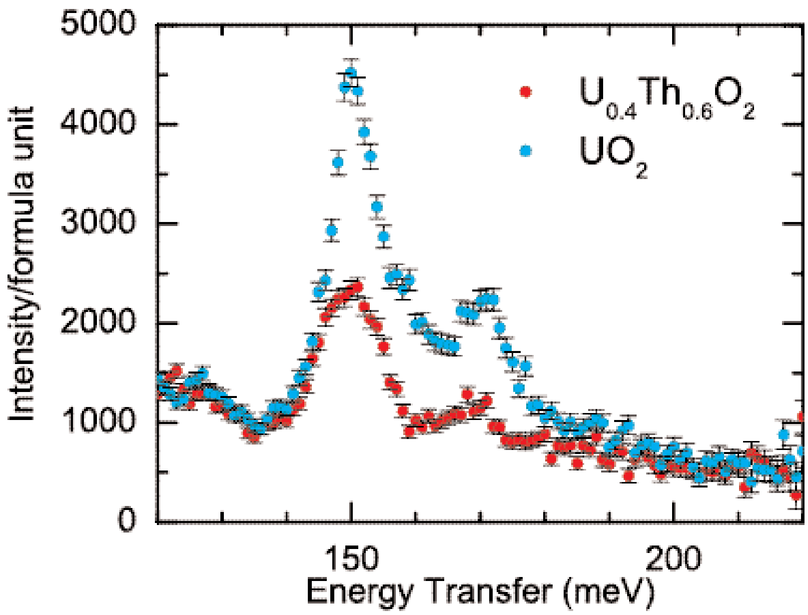
Magnon density of states in Fe_3O_4. Magnetite (Fe_3O_4) has been an important and largely unsolved problem in condensed-matter physics for 65 years. Magnetite is a ferrimagnet that undergoes a metal-insulator (Verwey) phase transition at 123 K. For many years, it has been assumed that the insulating ground state is charge-ordered, although that has been disputed recently.[3] Measurements on Pharos of the magnon density of states using Fe_3O_4 powders (Fig. 1) revealed changes in the magnon density-of-states through the Verwey transition. These measurements will help resolve the exchange energies and local ionic configurations in magnetite, and measurements of the magnon dispersion in single crystals are planned.

Spin-orbit excitations in UO_2. The crystal-field excitations of UO_2 have been of interest for the last 30 years. Following early attempts at the Intense Pulsed Neutron Source at Argonne National Laboratory,[4] important work was reported with higher resolution from ISIS (Rutherford-Appleton Laboratory Neutron Facility) in 1989, showing that the transitions from the ground-state/crystal-field state Γ_5 to excited states Γ_3 and Γ_4 were both in the



↑ Fig. 1. *(a) Low-angle (mainly magnetic) and high-angle (phonon) scattering from magnetite powder with Ei = 147 meV. Two clear spin-wave bands exist at 115 meV and 75 meV. (b) The 75-meV spin-wave band becomes broadened above the Verwey transition with little change in the 115-meV band.*

150- to 170-meV energy region.[5] The detailed spectra observed were modeled on the basis of both the exchange interactions and the Jahn-Teller (JT) effect caused by the distortion of the oxygen cage around the U atoms. Our experiments on UO_2 and a doped $(U_{0.4}Th_{0.6})O_2$ sample using Pharos shed important new light on the physics of these oxides. The doped compound does not order magnetically, so the exchange interactions are "switched off," leaving only the dynamic JT effect. Both the spectra from UO_2 for $T > T_N$ (30 K) (not shown) and from the doped compound show a broad first peak at 150 meV (Fig. 2). However, as shown in Fig. 2, the first peak from UO_2 for $T < T_N$ is sharp, which dramatically indicates the effect of the exchange field. Furthermore, the higher-energy peak at ~ 180 meV seen in the ISIS data, which cannot be reproduced by theory,[5] is not observed on Pharos, suggesting it may be spurious. Detailed modeling is under way.



↑ ***Fig. 2.*** *Inelastic scattering from polycrystalline samples of UO_2 (blue circles) and $(U_{0.4}Th_{0.6})O_2$ (red circles) both at 25 K. Both peaks are at the same position, but the pure compound has a much sharper spectrum. The incident energy was 243 meV, and the data are summed over the low-angle detectors.*

Conclusion

These early commissioning experiments show the potential of Pharos, and we are now starting the serious business of doing real science and proving the instrument. Proposals and interest in what we believe will be the best thermal beam direct-geometry chopper instrument in the U.S. are more than welcome. Moreover, Pharos will help build a user community ready for the SNS.

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